

Bounds on the mixing enhancement for a stirred binary fluid

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The Cahn–Hilliard equation describes phase separation in binary liquids. Here we study this equation with spatially-varying sources and stirring, or advection. We specialize to symmetric mixtures and time-independent sources and discuss stirring strategies that homogenize the binary fluid. By measuring fluctuations of the composition away from its mean value, we quantify the amount of homogenization achievable. We find upper and lower bounds on our measure of homogenization using only the Cahn–Hilliard equation and the incompressibility of the advecting flow. We compare these theoretical bounds with numerical simulations for two model flows: the constant flow, and the random-phase sine flow. Using the sine flow as an example, we show how our bounds on composition fluctuations provide a measure of the effectiveness of a given stirring protocol in homogenizing a phase-separating binary fluid.

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I. INTRODUCTION

Phase separation and its control have received intense interest, both because of industrial applications [1, 2, 3] and the mathematics involved, in particular the Cahn–Hilliard equation. This equation was introduced by Cahn and Hilliard [4] to describe phase separation in a binary alloy. Since their argument relies on the thermodynamic free energy of mixing, the equation is completely general, and describes any two-component system where the mixed state is energetically unfavorable, and where the total amount of matter is conserved. Thus it is used in polymer physics [2], in interfacial flows [5], and in mathematical biology [6]. In this paper we discuss binary liquids, and the order-parameter equation obtained by Cahn and Hilliard describes the composition of a binary fluid. A composition $c(\mathbf{x}, t) = 0$ indicates a locally well-mixed state, while a composition $c(\mathbf{x}, t) \neq 0$ indicates a local abundance of one binary fluid component over another. In the context of binary fluids, it is important to study the influence of stirring on the two fluid components, and we therefore introduce an advection term into the equation.

The time evolution of the non-advected equation has been studied extensively. In particular, there is a proof concerning the existence and uniqueness of solutions [7]. Given an initial state comprising a small perturbation around the unstable, well-mixed state $c = 0$, the system forms domains of unmixed fluid that expand or coarsen in time as $t^{1/3}$, the Lifshitz–Slyozov law [8, 9]. In many applications, the coarsening tendency of the Cahn–Hilliard fluid is undesirable, and this can be overcome by fluid advection, which is either

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passive or active. In the active case [10, 11, 12], the composition gradients induce a back-reaction on the flow, while in the passive case, the composition and its gradients exert no effect on the flow [13, 14, 15, 16], and it is this case that we consider in the present paper. If the flow contains large differential shears, the coarsening is either arrested, so that domains grow only to a certain size [13], or can be overwhelmed entirely, so that the mixed state, previously unstable, is now favored [15]. This homogenization is useful in applications, for example in the fabrication of emulsion paints [1].

Stirring provides one means of controlling phase separation. Indeed, it is well known that a unidirectional shear flow produces banded domains, with domains aligned in the shear direction [14, 17, 18, 19]. Other mechanisms have been employed to control the phase separation, including dipole interactions [20], patterned substrates [21], and surface forcing [22]. In [12], Ó Náráigh and Thiffeault study binary mixtures in thin films, and use the backreaction, together with surface forcing, to align the domains in a direction that depends on the forcing parameters. In the present paper, we focus on phase separation in the presence of advection and a spatially-varying persistent source. This source can be maintained in several ways. In [23] the spatial source is produced by thermal diffusion, through the Ludwig–Soret effect [24], in which composition gradients are induced by imposed temperature gradients. One can also produce composition gradients simply by injecting matter into the system [25]. In this paper, we shall restrict our attention to the symmetric binary fluid, in which equal amounts of both fluid components are present. This special case involves a wide range of phenomena [9, 11, 19, 23, 26]. In the present work, we shall refer to the combination of stirring (advection) and the spatial source as ‘forcing.’

We have mentioned the often undesirable coarsening tendency of the binary fluid, and the efforts made to suppress it. In this paper we introduce a quantitative measure of coarsening suppression by studying the p^{th} power-mean fluctuation of the composition about its average value. By fluctuations about the average value, we mean spatial fluctuations around the (constant) average spatial composition, which we then average over space and time. In effect, we study the time-averaged L^p norm of the composition. If this quantity is small, the average deviation of the system about the well-mixed state $c = 0$ is small, and we therefore use this quantity as a proxy for the level of mixedness of the fluid. An approach similar to this has already been taken for miscible fluids [27, 28, 29, 30, 31]. There, the equation of interest is the advection-diffusion equation, and fluctuations about the mean are measured by the variance or centred second power-mean of the fluid concentration [32, 33]. The variance is reduced by stirring. By specifying a source term, it is possible to state the maximum amount by which a given flow can reduce the variance, and hence mix the fluid. By quantifying the variance reduction, one can classify flows according to how effective they are at mixing. Just as the linearity of the advection-diffusion equation suggests the variance as a natural way of measuring fluctuations in the concentration, the non-linearity of the Cahn–Hilliard equation and its associated free energy (cubic and quartic in the composition, respectively), will fix our attention on the fourth power-mean of the composition fluctuations. Owing to Hölder’s inequality, a binary liquid that is well-mixed in this sense will also be well-mixed in the variance sense. The advantage we gain in considering the fourth power-mean is the derivation of explicit bounds on our measure of mixedness which have manifest flow- and source-dependence.

The paper is organized as follows. In Sec. II we introduce the model equations and discuss their nondimensionalization. We introduce measures of composition fluctuations and their relation to fluid mixing. These measures of composition fluctuations are based on long-

time averages obtained from the composition of the binary fluid. Therefore, in Sec. III we prove the existence of these long-time averages and find upper bounds on the measures of composition fluctuations. Since we are interested in minimizing composition fluctuations, in Sec. IV we obtain lower bounds on these measures. In Sec. V we investigate the parametric dependence of the upper and lower bounds for statistical, homogeneous, isotropic turbulence. In Sec. VI we compare the theoretical bounds with numerical simulations for two standard flows: the constant flow, and the sine flow. In the numerical simulations, we find that the composition fluctuations are indeed bounded by the theoretical limits we have obtained, and the results are dramatically different from those obtained for miscible liquids.

II. THE MODEL EQUATIONS

In this section, we introduce the advective Cahn–Hilliard equation with sources and discuss its properties. We outline the tools and notation we shall use to analyze composition fluctuations. For generality, the discussion takes place against the backdrop of scalar and vector fields in \mathbb{R}^n .

Let $c(\mathbf{x}, t)$ be the composition, that is, the scalar field $c(\mathbf{x}, t)$ measures phase separation, with $c(\mathbf{x}, t) = 0$ indicating a locally well-mixed state, and $c(\mathbf{x}, t) \neq 0$ indicating a local excess of one binary fluid component relative to the other. Let $\mathbf{v}(\mathbf{x}, t)$ be an externally imposed n -dimensional incompressible flow, $\nabla \cdot \mathbf{v} = 0$, and let $s(\mathbf{x})$ be a distribution of sources and sinks of binary fluid. The advective Cahn–Hilliard (ACH) equation describes the phase-separation dynamics of the scalar field $c(\mathbf{x}, t)$ in the presence of flow, for prescribed sources and sinks, is

$$\frac{\partial c}{\partial t} + \mathbf{v} \cdot \nabla c = D\Delta (c^3 - c - \gamma\Delta c) + s(\mathbf{x}). \quad (1)$$

Here D is Cahn–Hilliard diffusion coefficient and $\sqrt{\gamma}$ is the typical thickness of transition zones between phase-separated regions of the binary fluid. The finite thickness of these zones prevents the formation of infinite gradients in the problem. The equation is a passive advection equation: neither the composition nor its gradients affect the flow.

In this paper we work with a nondimensionalization of Eq. (1) that leaves three control parameters in the problem. Therefore, we can unambiguously study limits where control parameters tend to zero or infinity. Let T be a timescale associated with the velocity $\mathbf{v}(\mathbf{x}, t)$, and let V_0 be the magnitude of $\mathbf{v}(\mathbf{x}, t)$. Let S_0 be the magnitude of the source variations and, finally, let L be a lengthscale in the problem; for example, if the problem is solved in a cube with periodic boundary conditions, we take the lengthscale L to be the cube length. It is then possible to write down Eq. (1) using a nondimensional time $t' = t/T$ and a nondimensional spatial variable $\mathbf{x}' = \mathbf{x}/L$,

$$\frac{\partial c}{\partial t'} + V'_0 \tilde{\mathbf{v}} \cdot \nabla' c = D'\Delta' (c^3 - c - \gamma'\Delta' c) + S'_0 \tilde{s}(\mathbf{x}'),$$

$\tilde{\mathbf{v}}$ and \tilde{s} are dimensionless shape functions, $V'_0 = TV_0/L$, $D' = DT/L^2$, $\gamma' = \gamma/L^2$, and where $S'_0 = S_0T$. The quantity $D' = DT/L^2$ is identified with T/T_D , the ratio of the velocity timescale to the diffusion timescale. Following standard practice, we shall now work with the dimensionless version of the equation, and omit the prime notation. For ease of notation, we shall henceforth take \mathbf{v} to mean $V'_0\tilde{\mathbf{v}}$.

The equation (1) has the following properties, which we shall exploit in our analysis:

- If the source $s(\mathbf{x})$ is chosen to have spatial mean zero, then the total mass is conserved,

$$\frac{d}{dt} \int_{\Omega} c(\mathbf{x}, t) d^n x = \int_{\Omega} D \Delta (c^3 - c - \gamma \Delta c) d^n x + \int_{\Omega} s(\mathbf{x}) d^n x = 0 + [\text{boundary terms}],$$

where Ω is the problem domain in n dimensions and $|\Omega|$ is its volume. The boundary terms in this equation vanish on choosing no-flux boundary conditions $\hat{\mathbf{n}} \cdot \nabla c = \hat{\mathbf{n}} \cdot \nabla \mu = 0$ on $\partial\Omega$, or periodic boundary conditions. Here $\hat{\mathbf{n}} \cdot \nabla$ is the outward normal derivative. In this paper we shall consider the periodic case.

- There is a free-energy functional

$$F[c] = \int_{\Omega} \left[\frac{1}{4} (c^2 - 1)^2 + \frac{1}{2} \gamma |\nabla c|^2 \right] d^n x, \quad \mu = \frac{\delta F}{\delta c} = c^3 - c - \gamma \Delta c, \quad (2)$$

where μ is the chemical potential of the system [34]. For a smooth composition $c(\mathbf{x}, t)$, the free energy satisfies the evolution equation

$$\dot{F} \equiv \frac{dF}{dt} = -D \int_{\Omega} |\nabla \mu|^2 d^n x + \int_{\Omega} \mu (-\mathbf{v} \cdot \nabla c + s) d^n x,$$

and decays in time in the absence of sources and stirring.

To study the spatial fluctuations in composition, we consider the power-means of the quantity $c(\mathbf{x}, t) - |\Omega|^{-1} \int_{\Omega} c(\mathbf{x}, t) d^n x$,

$$M_p(t) = \left\{ \int_{\Omega} \left| c(\mathbf{x}, t) - \frac{1}{|\Omega|} \int_{\Omega} c(\mathbf{x}, t) d^n x \right|^p d^n x \right\}^{\frac{1}{p}}. \quad (3)$$

For a symmetric mixture in which $\int_{\Omega} c(\mathbf{x}, t) d^n x = 0$, this is simply

$$M_p(t) = \left\{ \int_{\Omega} |c(\mathbf{x}, t)|^p d^n x \right\}^{\frac{1}{p}} = \|c\|_p,$$

where we have introduced the L^p norm of the composition, $\|c\|_p$. The quantity M_p is a measure of the magnitude of spatial fluctuations in the composition about the mean, at a given time. Since we are interested in the ultimate state of the system, we study the long-time average of composition fluctuations. We therefore focus on the power-mean fluctuations

$$m_p = \langle M_p^p \rangle^{\frac{1}{p}},$$

where $\langle \cdot \rangle$ is the long-time average

$$\langle \cdot \rangle = \lim_{t \rightarrow \infty} \frac{1}{t} \int_0^t (\cdot) ds,$$

provided the limit exists. We shall repeatedly use the following results for the monotonicity of norms,

$$\begin{aligned} \|f\|_p &\leq |\Omega|^{\frac{1}{p} - \frac{1}{q}} \|f\|_q, & 1 \leq p \leq q, \quad f \in L^q(\Omega), \\ \frac{1}{t} \int_0^t |g(s)| ds &\leq \left[\frac{1}{t} \int_0^t |g(s)|^q ds \right]^{\frac{1}{q}}, & q \geq 1, \quad g \in L^s([0, t]), \end{aligned} \quad (4)$$

which follow from the Hölder inequality.

The Cahn–Hilliard equation and its free energy functional contain high powers of the composition c (c^3 and c^4 respectively), and we can therefore estimate m_p for specific p -values. In particular, in the following sections, we shall prove the following result in n dimensions:

Given a smooth solution to the ACH equation, the long-time average of the free energy exists, and therefore m_p exists, for $1 \leq p \leq 4$.

The constraints we impose on the forcing terms are that the velocity field and its first spatial derivatives be bounded in the L^∞ norm, and that the source term be bounded in the L^2 norm. That is, $\mathbf{v} \in L^\infty(0, T; H^{1,\infty}(\Omega))$ for any $T \in [0, \infty)$, and $s \in L^2(\Omega)$. We take our result one step further by explicitly evaluating upper and lower bounds for m_4 , and this gives a way of quantifying composition fluctuations in the stirred binary fluid.

III. EXISTENCE OF LONG-TIME AVERAGES

In this section, we prove a result concerning the existence of the long-time average of the free energy, and of the power-means m_p , for $1 \leq p \leq 4$.

Given the velocity field $\mathbf{v}(\mathbf{x}, t) \in L^\infty(0, T; H^{1,\infty}(\Omega))$ for any $T \in [0, \infty)$, the source $s(\mathbf{x}) \in L^2(\Omega)$, and smooth initial data for the ACH equation (1), the long-time average of the free energy exists, and thus m_p exists, for $1 \leq p \leq 4$.

The proof relies on the free-energy evolution equation. Using this law, we find uniform bounds on the finite-time means $\langle F \rangle_t$ and $\langle M_4^4 \rangle_t$, where

$$\langle \cdot \rangle_t = \frac{1}{t} \int_0^t (\cdot) ds, \quad \langle \cdot \rangle = \lim_{t \rightarrow \infty} \langle \cdot \rangle_t.$$

Using the monotonicity of norms, the uniform boundedness of $\langle M_p^p \rangle_t$, follows, for $1 \leq p \leq 4$. The proof proceeds in multiple steps, which we outline below.

Step 1: Analysis of the free-energy evolution equation

By modifying the argument of Elliott and Zheng [7] for the Cahn–Hilliard equation without flow and sources, it is readily seen that for smooth initial data, and for forcing terms with the regularity properties just mentioned, a unique smooth solution to the ACH equation exists, at least for finite times. Thus, we turn to the question of the long-time behaviour of solutions. We exploit the smoothness properties of the composition field $c(\mathbf{x}, t)$ and formulate an evolution equation for the free energy

$$F[c] = \int_\Omega \left[\frac{1}{4} (c^2 - 1) + \frac{1}{2} \gamma |\nabla c|^2 \right] d^n x.$$

Given the smooth, finite-time solution $c(\mathbf{x}, t)$, we differentiate the functional $F[c]$ with respect to time and obtain the relation

$$\frac{dF}{dt} = \int_\Omega \frac{\partial c}{\partial t} \mu d^n x, \quad \mu = c^3 - c - \gamma \Delta c.$$

Since $c(\mathbf{x}, t)$ satisfies the ACH equation (1), the evolution equation takes the form

$$\frac{dF}{dt} = -D \int_{\Omega} |\nabla \mu|^2 d^n x + \int_{\Omega} \mu (-\mathbf{v} \cdot \nabla c + s) d^n x,$$

using the no-flux or periodic boundary conditions. By averaging this equation over finite times, we obtain the identity

$$\langle \dot{F} \rangle_t + D \left\langle \int_{\Omega} |\nabla \mu|^2 d^n x \right\rangle_t = \left\langle \int_{\Omega} \mu s d^n x \right\rangle_t - \left\langle \int_{\Omega} \mu \mathbf{v} \cdot \nabla c d^n x \right\rangle_t, \quad (5)$$

We single out the quantity $\langle \dot{F} \rangle$ for study. Owing to the nonnegativity of $F(t)$, we have the inequality $\langle \dot{F} \rangle \geq 0$. Therefore, we need only consider two separate cases: $\langle \dot{F} \rangle = 0$, and $\langle \dot{F} \rangle > 0$. We shall show that $\langle \dot{F} \rangle \neq 0$ is not possible, and in doing so, we shall produce a uniform (t -independent) upper bound on $\langle F \rangle_t$.

Let us assume for contradiction that $\langle \dot{F} \rangle > 0$. Then, given any ε in the range $0 < \varepsilon < \langle \dot{F} \rangle$, there is a time T_ε such that $\langle \dot{F} \rangle - \varepsilon < \langle \dot{F} \rangle_t < \langle \dot{F} \rangle + \varepsilon$, for all times $t > T_\varepsilon$. Thus, for times $t > T_\varepsilon$, the time average $\langle F \rangle_t$ is strictly positive. Henceforth, the inequality $t > T_\varepsilon$ is assumed. We use the condition $\nabla \cdot \mathbf{v} = 0$, together with integration by parts, and find the last term in Eq. (5) becomes

$$\langle \dot{F} \rangle_t + D \left\langle \int_{\Omega} |\nabla \mu|^2 d^n x \right\rangle_t = \left\langle \int_{\Omega} \mu s d^n x \right\rangle_t + \left\langle \gamma \int_{\Omega} \Delta c \mathbf{v} \cdot \nabla c d^n x \right\rangle_t. \quad (6)$$

Now

$$\begin{aligned} \int_{\Omega} \Delta c \mathbf{v} \cdot \nabla c d^n x &= - \int_{\Omega} (\partial_i c) [\partial_i (v_j \partial_j c)] d^n x, \\ &= - \int_{\Omega} (\partial_i c) (\partial_i v_j) (\partial_j c) d^n x - \int_{\Omega} (\partial_i c) (\mathbf{v} \cdot \nabla) (\partial_i c) d^n x, \\ &= - \int_{\Omega} \mathbf{w} W \mathbf{w}^T d^n x, \end{aligned}$$

with

$$\mathbf{w} = \nabla c, \quad W_{ij} = \frac{1}{2} (\partial_i v_j + \partial_j v_i),$$

where we have used the summation convention for repeated indices and have omitted terms in the integration identities that vanish as a result of our choice of boundary conditions. The quadratic form $\mathbf{w} W \mathbf{w}^T$ satisfies $|\mathbf{w} W \mathbf{w}^T| \leq n \max_{ij} |W_{ij}| \|\mathbf{w}\|_2^2$, which gives rise to the inequality

$$\left| \int_{\Omega} \Delta c \mathbf{v} \cdot \nabla c d^n x \right| \leq n \left(\sup_{\Omega, i, j} |W_{ij}| \right) \int_{\Omega} |\nabla c|^2 d^n x. \quad (7)$$

The matrix W is the rate-of-strain tensor. The appearance of the rate-of-strain tensor in our analysis shows the importance of shear and stretching in the development of the composition morphology.

For each time $t' \in [0, t]$, we split the chemical potential μ into a part with mean zero, and a mean component: $\mu = \bar{\mu}(t') + \mu'(\mathbf{x}, t')$, where $\int_{\Omega} \mu'(\mathbf{x}, t') d^n x = 0$. Then, for any

function $\phi(\mathbf{x}, t)$ with spatial mean zero, we have the relation $\int_{\Omega} \phi \mu d^n x = \int_{\Omega} \phi \mu' d^n x$. Using this device, Eq. (6) becomes

$$\langle \dot{F} \rangle_t + D \left\langle \int_{\Omega} |\nabla \mu'|^2 d^n x \right\rangle_t = \left\langle \int_{\Omega} \mu' s d^n x \right\rangle_t + \left\langle \gamma \int_{\Omega} \Delta c \mathbf{v} \cdot \nabla c d^n x \right\rangle_t.$$

Owing to the positivity of $\langle \dot{F} \rangle_t$, we have the inequality

$$D \left\langle \int_{\Omega} |\nabla \mu'|^2 d^n x \right\rangle_t \leq \left\langle \int_{\Omega} \mu' s d^n x \right\rangle_t + \left\langle \gamma \int_{\Omega} \Delta c \mathbf{v} \cdot \nabla c d^n x \right\rangle_t. \quad (8)$$

Finally, we employ the Poincaré inequality for mean-zero functions on a periodic domain $\Omega = [0, L]^2$,

$$\|\mu'\|_2^2 \leq \left(\frac{L}{2\pi} \right)^2 \|\nabla \mu'\|_2^2. \quad (9)$$

Combining Eqs. (7), (8), and (9) gives the following inequality:

$$D \left(\frac{2\pi}{L} \right)^2 \langle \|\mu'\|_2^2 \rangle_t \leq \langle \|\mu'\|_2^2 \rangle_t^{\frac{1}{2}} \|s\|_2 + n W_{\infty} \left\langle \int_{\Omega} \gamma |\nabla c|^2 d^n x \right\rangle_t, \quad (10)$$

where $W_{\infty} = \sup_{t, \Omega, i, j} |W_{ij}|$. There are no angle brackets around the source term because $s(\mathbf{x})$ is independent of time.

Step 2: Obtaining a bound on $\langle \|\mu'\|_2^2 \rangle_t$

Using

$$\int_{\Omega} \gamma |\nabla c|^2 d^n x = \int_{\Omega} [\mu' c + c^2 - c^4] d^n x \leq \|\mu'\|_2 \|c\|_2 + \|c\|_2^2,$$

we obtain the inequality

$$\int_{\Omega} \gamma |\nabla c|^2 d^n x \leq |\Omega|^{\frac{1}{4}} \|\mu'\|_2 \|c\|_4 + |\Omega|^{\frac{1}{2}} \|c\|_4^2. \quad (11)$$

Combining Eqs. (10) and (11),

$$D \left(\frac{2\pi}{L} \right)^2 \langle \|\mu'\|_2^2 \rangle_t \leq \langle \|\mu'\|_2^2 \rangle_t^{\frac{1}{2}} \left[\|s\|_2 + n |\Omega|^{\frac{1}{4}} W_{\infty} \langle \|c\|_4^4 \rangle_t^{\frac{1}{4}} \right] + 2 |\Omega|^{\frac{1}{2}} W_{\infty} \langle \|c\|_4^4 \rangle_t^{\frac{1}{2}},$$

a quadratic inequality in $\langle \|\mu'\|_2^2 \rangle_t^{\frac{1}{2}}$. Hence,

$$\begin{aligned} \langle \|\mu'\|_2^2 \rangle_t^{\frac{1}{2}} &\leq \frac{1}{2D} \left(\frac{L}{2\pi} \right)^2 \left(\|s\|_2 + n |\Omega|^{\frac{1}{4}} W_{\infty} \langle \|c\|_4^4 \rangle_t^{\frac{1}{4}} \right) \\ &\quad + \frac{1}{2D} \left(\frac{L}{2\pi} \right)^2 \left[\left(\|s\|_2 + n |\Omega|^{\frac{1}{4}} W_{\infty} \langle \|c\|_4^4 \rangle_t^{\frac{1}{4}} \right)^2 + 8D |\Omega|^{\frac{1}{2}} \left(\frac{2\pi}{L} \right)^2 W_{\infty} \langle \|c\|_4^4 \rangle_t^{\frac{1}{2}} \right]^{\frac{1}{2}}. \end{aligned}$$

A less sharp bound is given by

$$\langle \|\mu'\|_2^2 \rangle_t \leq \frac{1}{D^2} \left(\frac{L}{2\pi} \right)^4 \left(\|s\|_2 + n |\Omega|^{\frac{1}{4}} W_{\infty} \langle \|c\|_4^4 \rangle_t^{\frac{1}{4}} \right)^2 + \frac{8 |\Omega|^{\frac{1}{2}}}{D} \left(\frac{L}{2\pi} \right)^2 W_{\infty} \langle \|c\|_4^4 \rangle_t^{\frac{1}{2}}, \quad (12)$$

which is an upper bound for $\langle \|\mu'\|_2^2 \rangle_t$, in terms of the forcing parameters and $\langle \|c\|_4^4 \rangle_t$.

Step 3: An upper bound on m_4

We have the free energy

$$\begin{aligned} F[c] &= \int_{\Omega} \left[\frac{1}{4} (c^2 - 1)^2 + \frac{1}{2} \gamma |\nabla c|^2 \right] d^n x = \int_{\Omega} \left[\frac{1}{2} c \mu - \frac{1}{4} c^4 \right] d^n x + \frac{1}{4} |\Omega| \\ &= \int_{\Omega} \left[\frac{1}{2} c \mu'(\mathbf{x}, t') - \frac{1}{4} c^4 \right] d^n x + \frac{1}{4} |\Omega| \geq 0. \end{aligned}$$

Hence,

$$\int_{\Omega} c^4 d^n x \leq 2 \int_{\Omega} c \mu' d^n x + |\Omega| \leq 2 \|c\|_2 \|\mu'\|_2 + |\Omega|.$$

Time-averaging both sides and using the monotonicity of norms (4), we obtain the result

$$\langle \|c\|_4^4 \rangle_t \leq |\Omega| + 2 |\Omega|^{\frac{1}{4}} \langle \|c\|_4^4 \rangle_t^{\frac{1}{4}} \langle \|\mu'\|_2^2 \rangle_t^{\frac{1}{2}}.$$

Using the bound for $\langle \|\mu'\|_2^2 \rangle_t$ in (12), this becomes

$$\begin{aligned} \langle \|c\|_4^4 \rangle_t &\leq |\Omega| \\ &+ \frac{2 |\Omega|^{\frac{1}{4}}}{D} \left(\frac{L}{2\pi} \right)^2 \langle \|c\|_4^4 \rangle_t^{\frac{1}{4}} \left[\left(\|s\|_2 + n |\Omega|^{\frac{1}{4}} W_{\infty} \langle \|c\|_4^4 \rangle_t^{\frac{1}{4}} \right)^2 + 4nD |\Omega|^{\frac{1}{2}} \left(\frac{2\pi}{L} \right)^2 W_{\infty} \langle \|c\|_4^4 \rangle_t^{\frac{1}{2}} \right]^{\frac{1}{2}}. \end{aligned}$$

We therefore have a t -independent equation for the upper bound on $\langle \|c\|_4^4 \rangle_t$,

$$\langle \|c\|_4^4 \rangle_t \leq m_4^{\max}(\mathbf{v}, s, D),$$

where m_4^{\max} solves the polynomial

$$\begin{aligned} (m_4^{\max})^4 &= |\Omega| \\ &+ \frac{2 |\Omega|^{\frac{1}{4}}}{D} \left(\frac{L}{2\pi} \right)^2 m_4^{\max} \left[\left(\|s\|_2 + n |\Omega|^{\frac{1}{4}} W_{\infty} m_4^{\max} \right)^2 + 4nD |\Omega|^{\frac{1}{2}} \left(\frac{2\pi}{L} \right)^2 W_{\infty} (m_4^{\max})^2 \right]^{\frac{1}{2}}, \end{aligned} \quad (13)$$

The highest power of m_4^{\max} on the left-hand side is $(m_4^{\max})^4$, while the highest power of m_4^{\max} on the right-hand side is $(m_4^{\max})^{\frac{3}{2}}$. Thus, this equation always has a unique positive solution.

We obtain the following chain of uniform (t -independent) bounds. Each bound follows from the previous bounds in the chain, and the first bound follows from Eq. (13).

- $\langle \|c\|_4^4 \rangle_t$ is uniformly bounded,
- $\langle \|c\|_2^2 \rangle_t$ is uniformly bounded,
- $\langle \|\mu'\|_2^2 \rangle_t$ is uniformly bounded, (14)
- $\langle \|\nabla c\|_2^2 \rangle_t$ is uniformly bounded,

- $\langle F \rangle_t$ is uniformly bounded,

for all $t > T_\varepsilon$. Owing to the uniformity of these bounds, they hold in the limit $t \rightarrow \infty$. The result $\langle F \rangle < \infty$ implies the existence of a uniform bound for $F(t)$, almost everywhere. Given the differentiability of $F(t)$, this implies that $F(t)$ is everywhere uniformly bounded, and thus, $\langle \dot{F} \rangle = 0$, which is a contradiction. Therefore, the only possibility for $\langle F \rangle$ is that it be zero. It is straightforward to verify that by taking $\langle \dot{F} \rangle = 0$, and making slight alterations in steps 1–3, the bounds in Eq. (14) still hold.

Let us examine the significance of our result. We have shown that for sufficiently regular flows and source terms (specifically, $\mathbf{v}(\mathbf{x}, t) \in L^\infty(0, T; H^{1,\infty}(\Omega))$, $T \in [0, \infty)$, and $s(\mathbf{x}) \in L^2(\Omega)$), there is an *a priori* bound on the compositional free energy $\langle F[c] \rangle$. We have shown that the system always reaches a steady state, in the sense that $\langle \dot{F} \rangle = 0$. We have also found an upper bound for the m_4 measure of composition fluctuations, as the unique positive root of the polynomial equation Eq. (13). This bound depends only on the source amplitude, the diffusion constant, and the maximum rate-of-strain W_∞ . Using the monotonicity of norms, this number serves also as an upper bound on m_p for $1 \leq p \leq 4$. Let us comment briefly on the volume term in the equation $(m_4^{\max})^4 = |\Omega| + \dots$. Since this upper bound includes many situations, it must take into account the case where both the velocity and the source vanish. Then $c \sim \pm 1$ as $t \rightarrow \infty$, and by definition, $m_4 \sim |\Omega|^{\frac{1}{4}}$, which is in agreement with Eq. (13).

As mentioned in Sec. I, it is desirable in many applications to suppress composition fluctuations, since this leads to a homogeneous mixture. In this paper, we propose advection as a suppression mechanism, and we would therefore like to know the maximum suppression achievable for a given flow. This suggests that we seek lower bounds on m_p , in addition to the upper bounds found in this section.

IV. LOWER BOUNDS ON THE COMPOSITION FLUCTUATIONS

In this section we discuss the significance of the lower bound on the measure m_p of composition fluctuations. Due to the powers of the composition that appear in the Cahn–Hilliard equation, it is possible to obtain an explicit lower bound for m_4 , which we then use to discuss mechanisms to suppress composition fluctuations. After taking care of the volume factors, the lower bound on m_4 must be greater than or equal to the lower bound on m_p , for $1 \leq p \leq 4$. Thus, a flow that suppresses composition fluctuations in the m_4 sense will also suppress them in the m_p sense, for $1 \leq p \leq 4$.

As discussed in Sec. II, a suitable measure of composition fluctuations for a symmetric mixture is

$$m_p = \langle \|c\|_p^p \rangle^{\frac{1}{p}},$$

where $c(\mathbf{x}, t)$ is the composition of the binary mixture and $\|c\|_p$ is its L^p norm. For $p = 2$, this gives the usual variance, used in the theory of miscible fluid mixing [27, 28, 29, 30, 31]. In that case, the choice $p = 2$ is a natural one suggested by the linearity of the advection–diffusion equation. In the following analysis of the ACH equation, it is possible to find an explicit lower bound for m_4 and we therefore use this quantity to study the suppression of composition fluctuations due to the imposed velocity field. Given this formula, we can compare the suppression achieved by a given flow with the ideal level of suppression, and decide on the best strategy to homogenize the binary fluid.

To estimate m_4 , we take the ACH equation (1), multiply it by an arbitrary, spatially-varying test function $\phi(\mathbf{x})$, and then integrate over space and time, which yields

$$\left\langle \int_{\Omega} c [\hat{Q}\phi + Dc^2\Delta\phi] d^n x \right\rangle = - \int_{\Omega} s\phi d^n x, \quad (15)$$

where \hat{Q} is the linear operator $\mathbf{v} \cdot \nabla - D\Delta - \gamma D\Delta^2$. Using the constraint Eq. (15), the monotonicity of norms, and the Cauchy–Schwarz inequality, we obtain the following string of inequalities,

$$\left| \int_{\Omega} s\phi d^n x \right| \leq \langle \|c\|_2 \|\hat{Q}\phi + Dc^2\Delta\phi\|_2 \rangle \leq \langle \|c\|_2^2 \rangle^{\frac{1}{2}} \langle \|\hat{Q}\phi + Dc^2\Delta\phi\|_2^2 \rangle^{\frac{1}{2}},$$

which gives the relation

$$\langle \|c\|_2^2 \rangle^{\frac{1}{2}} \geq \frac{\left| \int_{\Omega} s\phi d^n x \right|}{\langle \|\hat{Q}\phi + Dc^2\Delta\phi\|_2^2 \rangle^{\frac{1}{2}}}.$$

We study the denominator

$$\begin{aligned} \langle \|\hat{Q}\phi + Dc^2\Delta\phi\|_2^2 \rangle^{\frac{1}{2}} &\leq \langle \|\hat{Q}\phi\|_2^2 \rangle^{\frac{1}{2}} + D \langle \|c^2\Delta\phi\|_2^2 \rangle^{\frac{1}{2}}, \\ &\leq \left\langle \int_{\Omega} (\hat{Q}\phi)^2 d^n x \right\rangle^{\frac{1}{2}} + D \|\Delta\phi\|_{\infty} \langle \|c\|_4^4 \rangle^{\frac{1}{2}}, \end{aligned}$$

where this bound follows from the triangle and Hölder inequalities. Thus we have the result

$$\langle \|c\|_2^2 \rangle^{\frac{1}{2}} \geq \frac{\left| \int_{\Omega} s\phi d^n x \right|}{\left\langle \int_{\Omega} (\hat{Q}\phi)^2 d^n x \right\rangle^{\frac{1}{2}} + D \|\Delta\phi\|_{\infty} \langle \|c\|_4^4 \rangle^{\frac{1}{2}}},$$

or

$$\langle \|c\|_2^2 \rangle^{\frac{1}{2}} \left[\left\langle \int_{\Omega} (\hat{Q}\phi)^2 d^n x \right\rangle^{\frac{1}{2}} + D \|\Delta\phi\|_{\infty} \langle \|c\|_4^4 \rangle^{\frac{1}{2}} \right] \geq \left| \int_{\Omega} s\phi d^n x \right|.$$

Using the monotonicity of norms (4), we recast this inequality as one involving only a single power-mean,

$$|\Omega|^{\frac{1}{4}} \langle \|c\|_4^4 \rangle^{\frac{1}{4}} \left[\left\langle \int_{\Omega} (\hat{Q}\phi)^2 d^n x \right\rangle^{\frac{1}{2}} + D \|\Delta\phi\|_{\infty} \langle \|c\|_4^4 \rangle^{\frac{1}{2}} \right] \geq \left| \int_{\Omega} s\phi d^n x \right|.$$

Therefore, we have the following inequality for $m_4 = \langle \|c\|_4^4 \rangle^{\frac{1}{4}}$,

$$m_4 (q_0(\mathbf{v}, D, \gamma) + D \|\Delta\phi\|_{\infty} m_4^2) \geq |\Omega|^{-\frac{1}{4}} \left| \int_{\Omega} s\phi d^n x \right|,$$

where

$$q_0(\mathbf{v}, D, \gamma) = \left\langle \int_{\Omega} [\mathbf{v} \cdot \nabla \phi - D\Delta\phi - D\gamma\Delta^2\phi]^2 d^n x \right\rangle^{\frac{1}{2}}.$$

Thus, we obtain a lower bound for the m_4 measure of composition fluctuations,

$$m_4 \geq m_4^{\min}, \quad D \|\Delta\phi\|_{\infty} (m_4^{\min})^3 + q_0(\mathbf{v}, s, D) m_4^{\min} - |\Omega|^{-\frac{1}{4}} \left| \int_{\Omega} s\phi d^n x \right| = 0. \quad (16)$$

The cubic equation satisfied by m_4^{\min} has a unique positive root.

To probe the asymptotic forms of (16), we rewrite the forcing terms $\mathbf{v}(\mathbf{x}, t)$ and $s(\mathbf{x})$ as an amplitude, multiplied by a dimensionless shape function. Thus,

$$\begin{aligned}\mathbf{v} &= V_0 \tilde{\mathbf{v}}, & V_0 &= |\Omega|^{-\frac{1}{2}} \langle \|\mathbf{v}\|_2^2 \rangle^{\frac{1}{2}}, \\ s &= S_0 \tilde{s}, & S_0 &= |\Omega|^{-\frac{1}{2}} \|s\|_2,\end{aligned}$$

Then for a fixed value of S_0 and D , and $V_0 \gg 1$ (large stirring), we have $q_0 \sim V_0 \langle \int (\tilde{\mathbf{v}} \cdot \nabla \phi)^2 d^n x \rangle^{\frac{1}{2}}$, and the lower bound m_4^{\min} takes the form

$$m_4^{\min} \sim \frac{S_0}{V_0} \frac{|\int_{\Omega} \tilde{s} \phi d^n x|}{\langle \int_{\Omega} (\tilde{\mathbf{v}} \cdot \nabla \phi)^2 d^n x \rangle^{\frac{1}{2}} |\Omega|^{\frac{1}{4}}}, \quad V_0 \gg 1.$$

On the other hand, for fixed S_0 and V_0 , and $D \gg 1$ (large diffusion), the lower bound takes the form

$$m_4^{\min} \sim \frac{S_0}{D} \frac{|\langle \int_{\Omega} \tilde{s} \phi d^n x \rangle|}{[\int_{\Omega} (\Delta \phi + \gamma \Delta^2 \phi)^2 d^n x]^{\frac{1}{2}} |\Omega|^{\frac{1}{4}}}, \quad D \gg 1.$$

It is possible to obtain similar asymptotic expressions for m_2 , by constrained minimization of a functional of the composition. Apart from a volume factor, the asymptotic form of m_2 agrees exactly with the asymptotic form of m_4 just obtained. The functional to be minimized is

$$\Phi[c] = \frac{1}{2} \left\langle \int_{\Omega} c^2 d^n x \right\rangle - \lambda \left\langle \int_{\Omega} (c \hat{Q} \phi + D c^3 \Delta \phi + s \phi) d^n x \right\rangle,$$

where $\phi(\mathbf{x})$ is a test function. (This approach was used in [30] for the advection-diffusion equation.) Setting $\delta \Phi / \delta c = 0$ gives

$$c = \frac{1 - \sqrt{1 - 12\lambda^2 D \Delta \phi \hat{Q} \phi}}{6\lambda D \Delta \phi}. \quad (17)$$

Evaluation of $\delta^2 \Phi / \delta c \delta c'$ shows that Eq. (17) produces a minimum of $\Phi[c]$. Given the expression $\hat{Q} = V_0 \tilde{\mathbf{v}} \cdot \nabla - D \Delta - \gamma D \Delta^2$, the minimum Eq. (17) is $\lambda V_0 \tilde{\mathbf{v}} \cdot \nabla \phi$ at large V_0 . Substitution of this expression into the constraint $\langle \int_{\Omega} [c \hat{Q} \phi + D c^3 \Delta \phi + s \phi] d^n x \rangle = 0$ gives $\lambda = -(S_0/V_0^2) [\langle \int_{\Omega} \tilde{s} \phi d^n x \rangle / \langle \int_{\Omega} (\tilde{\mathbf{v}} \cdot \nabla \phi)^2 d^n x \rangle]$, and hence

$$m_2^{\min} \sim \frac{S_0}{V_0} \frac{|\langle \int_{\Omega} \tilde{s} \phi d^n x \rangle|}{\langle \int_{\Omega} (\tilde{\mathbf{v}} \cdot \nabla \phi)^2 d^n x \rangle^{\frac{1}{2}}}, \quad V_0 \gg 1.$$

For fixed V_0 and S_0 and large D , a similar calculation gives

$$m_2^{\min} \sim \frac{S_0}{D} \frac{|\langle \int_{\Omega} \tilde{s} \phi d^n x \rangle|}{[\int_{\Omega} (\Delta \phi + \gamma \Delta^2 \phi)^2 d^n x]^{\frac{1}{2}}}, \quad D \gg 1.$$

These expressions show that, apart from a volume factor, the lower bounds on the m_2 and m_4 measures of composition fluctuations are identical in the limits of high stirring strength or high diffusion. In particular, the asymptotic expression

$$m_2^{\min}, \quad |\Omega|^{\frac{1}{4}} m_4^{\min} \sim \frac{S_0}{V_0} \frac{|\int_{\Omega} \tilde{s} \phi d^n x|}{\langle \int_{\Omega} (\tilde{\mathbf{v}} \cdot \nabla \phi)^2 d^n x \rangle^{\frac{1}{2}}}, \quad \text{for large } V_0,$$

indicates that if a flow can be found that saturates the lower bound $m_{2,4}^{\min}$, the suppression of composition fluctuations can be enhanced by a factor of V_0^{-1} at large stirring amplitudes. Such a flow would then be an efficient way of mixing the binary fluid.

V. SCALING LAWS FOR m_4

In this section we investigate the dependence of the m_4 measure of composition fluctuations on the parameters of the problem, namely the stirring velocity \mathbf{v} , the source s , and the diffusion constant D . For simplicity, we shall restrict our interest to a certain class of flows, which enables us to compute long-time averages explicitly.

The lower bound for the m_4 measure of composition fluctuations is the unique positive root of the polynomial

$$D\|\Delta\phi\|_\infty (m_4^{\min})^3 + q_0(\mathbf{v}, D, \gamma) m_4^{\min} - |\Omega|^{-\frac{1}{4}} \left| \int_\Omega s\phi d^n x \right| = 0, \quad (18)$$

where $\phi(\mathbf{x})$ is a test function and

$$q_0(\mathbf{v}, D, \gamma) = \left\langle \int_\Omega [\mathbf{v} \cdot \nabla\phi - D\Delta\phi - D\gamma\Delta^2\phi]^2 d^n x \right\rangle^{\frac{1}{2}}.$$

Following [27, 28], we specialize to velocity fields whose time average has the following properties,

$$\langle v_i(\mathbf{x}, \cdot) \rangle = 0, \quad \langle v_i(\mathbf{x}, \cdot) v_j(\mathbf{x}, \cdot) \rangle = \frac{V_0^2}{n} \delta_{ij}. \quad (19)$$

The flow $\mathbf{v}(\mathbf{x}, t)$ is defined on the n -torus $[0, L]^n$. A statistically homogeneous and isotropic turbulent velocity field automatically satisfies the relations (19), although it is not necessary for \mathbf{v} to be of this type. The source we consider is monochromatic (that is, it contains a single spatial scale) and varies in a single direction,

$$s = \sqrt{2}S_0 \sin(k_s x). \quad (20)$$

Our choice of velocity field makes the evaluation of $q_0(\mathbf{v}, D, \gamma)$ particularly easy:

$$q_0(\mathbf{v}, D, \gamma) = \left[\frac{V_0^2}{n} \int_{[0,L]^n} |\nabla\phi|^2 d^n x + D^2 \int_{[0,L]^n} (\Delta\phi + \gamma\Delta^2\phi)^2 d^n x \right]^{\frac{1}{2}}.$$

In studies of the advection-diffusion equation [30], it is possible to find an explicit test function ϕ that sharpens the lower bound on m_2 . The procedure for doing this depends on the linearity of the equation. Here, this is not possible, and for simplicity we set $\phi = s$. This choice of ϕ certainly gives a lower bound for the m_4 measure of composition fluctuations, with the added advantage of enabling explicit computations. Having specified the coefficients of the polynomial in Eq. (18) completely, we extract the positive root of this equation, and find the lower bound m_4^{\max} , as a function of V_0 . The results of this procedure are shown in Fig. 1.

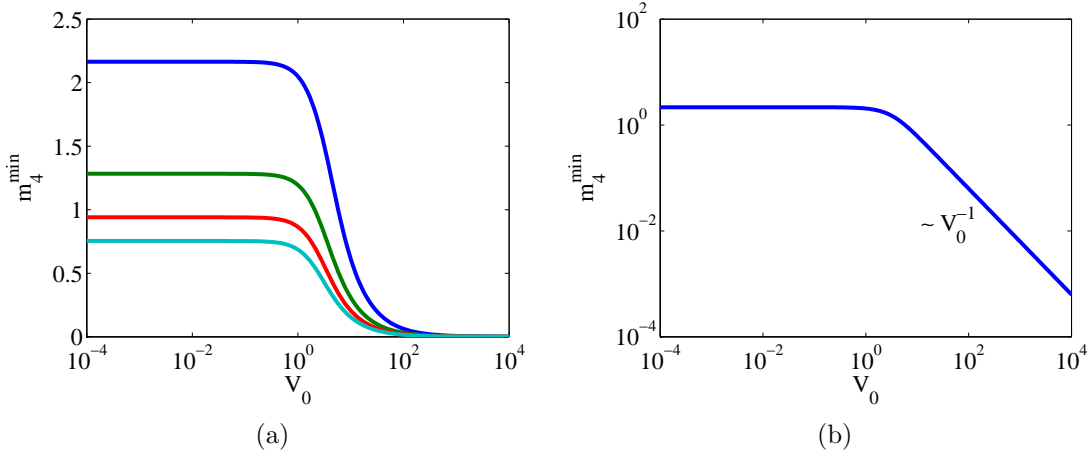


FIG. 1: (a) The lower bound for m_4^{\min} as a function of V_0 for a monochromatic source; (b) the dependence of m_4^{\min} on the velocity amplitude V_0 . In (a), the scale of the source variation decreases in integer multiples from $k_s = 2\pi/L$ in the uppermost curve, to $k_s = 8\pi/L$ in the lowermost curve, while in (b) the source scale is set to $2\pi/L$. In both figures, we have set $D = S_0 = 1$.

Let us examine briefly the scaling of the upper bound m_4^{\max} with the problem parameters. The upper bound satisfies the polynomial equation

$$(m_4^{\max})^4 = |\Omega| + \frac{2|\Omega|^{\frac{1}{4}}}{D} \left(\frac{L}{2\pi}\right)^2 m_4^{\max} \left[\left(S_0 |\Omega|^{\frac{1}{2}} + n |\Omega|^{\frac{1}{4}} W_\infty m_4^{\max} \right)^2 + 4nD |\Omega|^{\frac{1}{2}} \left(\frac{2\pi}{L}\right)^2 W_\infty (m_4^{\max})^2 \right]^{\frac{1}{2}}, \quad (21)$$

which depends only on the diffusion D , the source amplitude S_0 , and the maximum rate-of-strain W_∞ . For W_∞ large, the flow-dependence of the upper bound is $m_4^{\max} \sim W_\infty^{\frac{1}{2}}$. This dependence is verified by obtaining the positive root of Eq. (21), which is a function of W_∞ . The results are shown in Fig. 2.

In this section we have investigated the parametric dependence of the theoretical bounds $m_4^{\max, \min}$, for flows with the properties in Eq. (19). We note that for any nonzero source, the lower bound m_4^{\min} is nonzero, meaning that no matter how hard one stirs, there will always be some inhomogeneity in the fluid, and this is in fact true for any flow. However, the number m_4^{\min} tells us how much homogeneity we can achieve and is therefore a yardstick for stirring protocols. We use this yardstick to test model flows in the next section.

VI. NUMERICAL SIMULATIONS

In this section we solve Eq. (1) numerically for two flows, and verify the bounds obtained in Secs. III–V. We use the sinusoidal source term in Eq. (20) with periodic boundary conditions, and the source scale k_s therefore takes the form $(2\pi/L)j$, where L is the box size and j is an integer. We specialize to two dimensions and study two standard flows that are used in the analysis of mixing: the random-phase sine flow [25, 29, 35, 36, 37], and the constant flow [31].

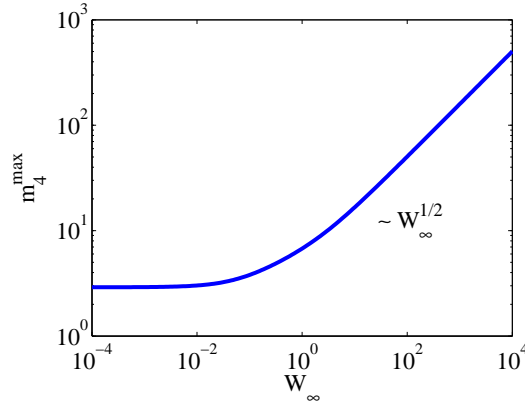


FIG. 2: The dependence of the upper bound m_4^{\max} on the maximum rate-of-strain W_∞ . The source affects the upper bound only through its square mean $S_0 = |\Omega|^{-\frac{1}{2}} \|s\|_2$.

Random-phase sine flow

The random-phase sine flow is the time-dependent two-dimensional flow

$$\begin{aligned} v_x(x, y, t) &= \sqrt{2} V_0 \sin(k_v y + \phi_j), & v_y &= 0, & j\tau \leq t < (j + \tfrac{1}{2})\tau, \\ v_y(x, y, t) &= \sqrt{2} V_0 \sin(k_v x + \psi_j), & v_x &= 0, & (j + \tfrac{1}{2})\tau \leq t < (j + 1)\tau, \end{aligned} \quad (22)$$

where ϕ_j and ψ_j are phases that are randomized once during each flow period τ , and where the integer j labels the period. The flow is defined on the two-dimensional torus $[0, L]^2$. The time average of this velocity field has the properties listed in Eq. (19). Because of its simplicity, the sine flow is a popular testbed for studying chaotic mixing [25, 29, 35, 36, 37].

We solve Eq. (1) with the flow in Eq. (22) using an operator splitting: the advection step is carried out using the lattice method of Pierrehumbert [25, 37], and the subsequent Cahn–Hilliard and source steps are implemented simultaneously using a spectral method [9]. The nondimensionalization outlined in Sec. II is appropriate here: the unit of time T is identified with the flow period τ , and the unit of length is the box size L . The control parameters in the problem are the dimensionless velocity V_0 , the dimensionless diffusion D , and the dimensionless source amplitude $S_0 = |\Omega|^{-\frac{1}{2}} \|s\|_2$. We use V_0 as a measure of stirring intensity and fix the other parameters in what follows. The flow we choose is chaotic at all stirring amplitudes and given our choice of scaling, has Lyapunov exponent [15]

$$\lambda \sim 0.236 V_0^2, \quad V_0 \ll 1; \quad \lambda \sim \log\left(\frac{V_0^2}{2}\right), \quad V_0 \gg 1.$$

The lattice method with its splitting of the advection and diffusion steps, is effective only when diffusion is slow compared to advection, that is, $T/T_D \ll 1$. We therefore set $D = 10^{-5}$, with $\tau = L = 1$. A numerical experiment with $V_0 = 0$ shows that $S_0 = 5 \times 10^{-4}$ gives rise to a morphology that is qualitatively different from the sourceless case, and we therefore work with this source amplitude. Finally, following standard practice [11, 13], we choose $\gamma \sim \Delta x^2$, the gridsize.

Using these new scaling rules, and the identity $W_\infty = \sqrt{2} V_0 k_v$ for the sine flow, we recall

the large-stirring forms of $m_4^{\max, \min}$, For $V_0 \gg D$, the lower bound has the form

$$m_4^{\min} \sim \frac{S_0}{V_0} \frac{\int_{\Omega} \sin(k_s x) \phi d^2 x}{[\int_{\Omega} |\nabla \phi|^2 d^2 x]^{\frac{1}{2}}}, \quad (23)$$

with the power-law relationship $m_4^{\min} \sim V_0^{-1}$, while for small $V_0 \gg 1$ the upper bound has the form

$$m_4^{\max} \sim \left(\frac{2^{\frac{1}{2}} k_v}{\pi^2 D} \right)^{\frac{1}{2}} V_0^{\frac{1}{2}}. \quad (24)$$

These scaling results are identical to those for the advection-diffusion problem [29].

Before studying the case with flow, we integrate Eq. (1) without flow, to verify the effect of the source. For a sufficiently large source amplitude, the composition phase-separates

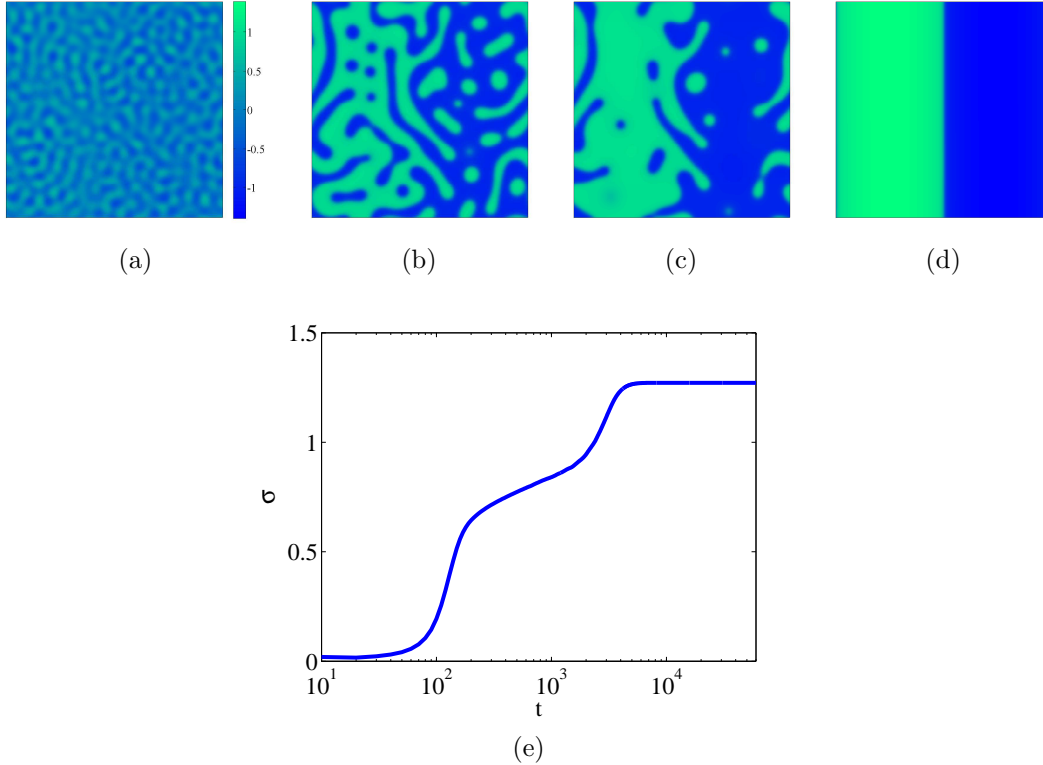


FIG. 3: The composition of the binary fluid for $S_0 = 5 \times 10^{-4}$ and (a) $t = 100$; (b) $t = 1000$; (c) $t = 2000$; (d) $t = 8000$. A steady state is reached in (d), evidenced by the time dependence of m_4 in (e), where m_4 is constant for $t \gg 1$.

and forms domains rich in either binary fluid component. These domains are aligned with variations in the source. A steady-state is reached and m_4 attains a constant value, as seen in Fig. 3. On the other hand, for small source amplitudes, we have verified that the domains do not align with the source, and their growth does not saturate. We do not consider this case here, since we are interested in sources that qualitatively alter the phase separation. These different regimes are discussed in [23].

We consider the case with flow by varying V_0 , and find results that are similar to those found in [15], for the same stirring mechanism without sources. For all values of V_0 , the

composition reaches a steady state, in which $\|c\|_4$, the pre-averaged form of m_4 , fluctuates around a constant value. For small values of V_0 , the domain growth is arrested due to a balance between the advection and phase-separation terms in the equation, while for moderate values of V_0 , the domains are broken up and a mixed state is obtained. At large values of V_0 , the m_4 measure of composition fluctuations saturates: further increases in V_0 do not produce further decreases in m_4 . At these large values of V_0 , the source structure is visible in snapshots of the composition, as evidenced by Fig. 4.

We investigate the dependence of composition fluctuations on the stirring strength V_0 , and show the results in Fig. 5. The theoretical upper and lower bounds on m_4 depend on V_0 and are obtained as roots of Eqs. (18) and (21). In the limit of large V_0 , these bounds have power-law behaviour, with $m_4^{\max} \sim V_0^{\frac{1}{2}}$ and $m_4^{\min} \sim V_0^{-1}$, as demonstrated by Eqs. (23) and (24). The numerical values of m_4 are indeed bounded by these limiting values, although the V_0 -dependence is not a power law. Instead, the function $m_4(V_0)$ is a non-increasing

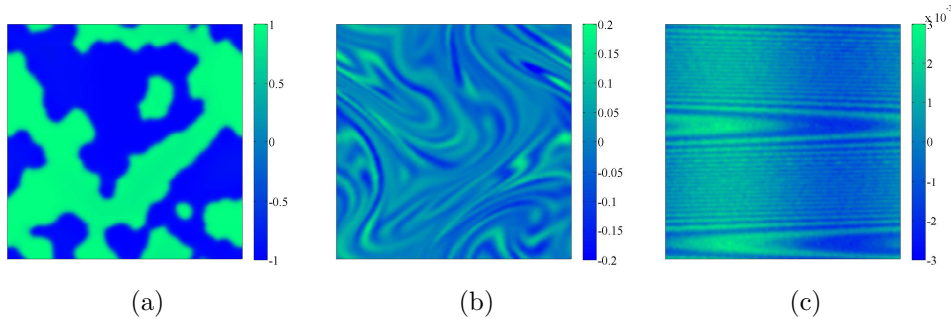


FIG. 4: A snapshot of the steady-state composition for (a) $V_0 = 0.001$; (b) $V_0 = 0.1$; (c) $V_0 = 10$. In (a) domain growth is arrested, in (b) the domains are destroyed and the binary fluid mixes, while in (c) m_4 measure of composition fluctuations is minimized, and the source structure is visible.

function, with a sharp drop occurring in a small range of V_0 -values. Thus, the fluid becomes more homogeneous with increasing V_0 . We discuss the effect of stirring on the inhomogeneity of the fluid by introducing the notion of mixing enhancement.

We measure the ability of a given stirring protocol to suppress composition fluctuations by the mixing enhancement. Similar ideas are often applied to the advection-diffusion equation [27, 28, 29, 30, 31]. We define the dimensionless mixing enhancement

$$\eta_p \equiv \frac{m_p^{\min}(V_0 = 0)}{m_p(V_0)}.$$

For a given flow, the number η_p quantifies the flow's ability to suppress composition fluctuations. In a well-mixed flow, the local deviation of $c(\mathbf{x}, t)$ away from the mean will be small; a small m_p -value is a signature of a well-mixed flow. We are therefore justified in calling η_p the mixing enhancement. We obtain some control over the mixing enhancement η_4 from the inequalities of Secs. III and IV. Based on these inequalities, the mixing enhancement is bounded above and below,

$$\eta_4^{\min} \equiv \frac{m_4^{\min}(V_0 = 0)}{m_4^{\max}(V_0)} \leq \eta_4 \leq \frac{m_4^{\min}(V_0 = 0)}{m_4^{\min}(V_0)} \equiv \eta_4^{\max}.$$

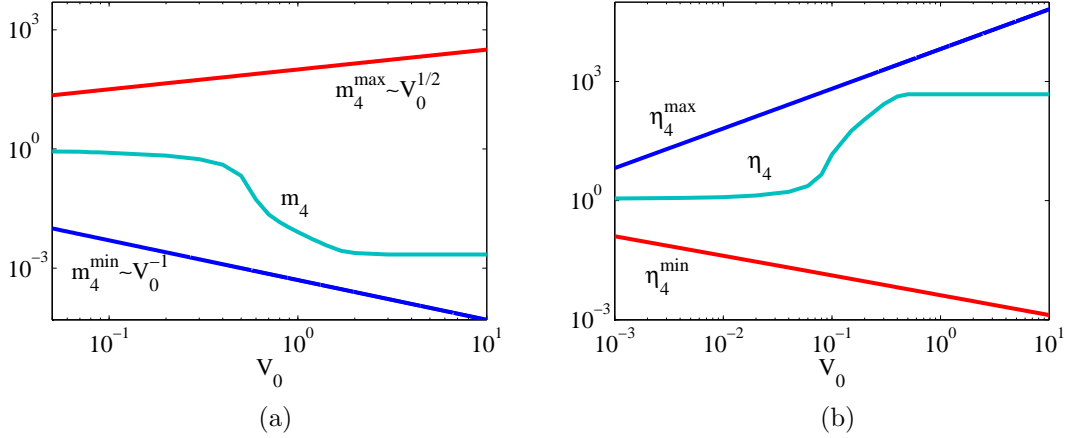


FIG. 5: (a) The m_4 measure of composition fluctuations or mixing for the sine flow, as a function of the stirring parameter V_0 . The values of D and S_0 are given in the text. The upper and lower bounds are shown for comparison; (b) The mixing enhancement η_4 for the sine flow, with the upper and lower bounds shown for comparison.

We have plotted the upper and lower bounds on the mixing enhancement for the case of monochromatic sources in Fig. 5(b). The maximum enhancement always exceeds unity in this case, which implies the possibility of finding stirring protocols that homogenize the fluid. On the other hand, the minimum enhancement is less than unity, which indicates the possibility of finding stirring protocols that actually amplify composition fluctuations, and this amplification depends weakly on the maximum rate-of-strain W_∞ . This latter case is not surprising, given that a uniform shear flow causes the domains of the Cahn–Hilliard fluid to align, rather than to break up. The sine-flow enhancement is a non-decreasing function of the stirring parameter V_0 . At small values of V_0 , small increases in the vigor of stirring lead to small increases in the mixing enhancement. There is a window of intermediate V_0 -values for which the mixing enhancement increases sharply with increasing V_0 . At higher values of V_0 , the efficiency saturates, so that further increases in the vigor of stirring have no effect on composition fluctuations. The saturation is due to finite-size effects: the sine flow wraps filaments of fluid around the torus as in Fig. 4(c).

Constant flow

We study the flow $(v_x, v_y) = (0, V_0)$, where V_0 is a constant. We choose a nondimensionalization that is set by the diffusion time $T_D = L^2/D$, and obtain the following parametric version of Eq. (1),

$$\frac{\partial c}{\partial t'} + V_0' \frac{\partial c}{\partial y'} = \Delta' (c^3 - c - \gamma' \Delta' c) + \sqrt{2} S_0' \sin(k_s' x'), \quad (25)$$

where $V_0' = LV_0/D$, $\gamma' = \gamma/L^2$, and $S_0' = S_0 L^2/D$. We immediately drop the primes from Eq. (25). We fix γ and S_0 and vary the flow strength V_0 . This flow does not satisfy the time-correlation relations (19), although the maximum rate-of-strain has the simple form $W_\infty = 0$. Using this information, and the test function $\phi = s(x)$, the upper and lower

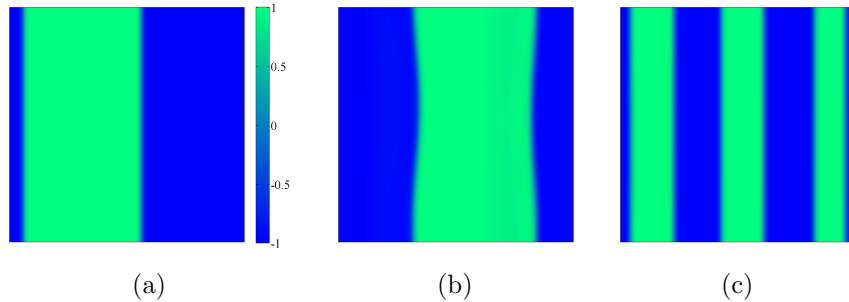


FIG. 6: A snapshot of the steady-state composition for (a) $V_0 = 10$; (b) $V_0 = 100$; (c) $V_0 = 1000$.

bounds obtained in Eqs. (13) and (16) are independent of the flow strength.

We solve Eq. (25) numerically for various values of V_0 and present the results in Fig. 6. For small V_0 , the composition morphology is similar to the flowless case seen in Fig. 3(d), except now the domains are uniformly advected in a direction perpendicular to the source variation. The small- V_0 case is shown in Fig. 6(a). As V_0 increases, the domain boundaries are distorted due to the advection, while for large V_0 , the advection is sufficiently strong to break up the laminar domains. The domain-like structure persists at late times however, and narrower laminar domains form.

The m_4 measure of mixedness is almost constant across the range of stirring parameters $0 \leq V_0 \leq 1000$. For $V_0 = 1000$, m_4 is slightly smaller than its value at $V_0 = 0$, due to the presence of more interfaces. This difference is small however, and increasing V_0 does little to mix the fluid. This is not surprising, given that local shears are necessary to break up domain structures [11], and that such shears are absent from constant flows. What this example shows however, is the difference between a miscible mixture, and a phase-separating mixture. For a diffusive mixture with the sinusoidal source we have studied, the constant flow discussed here is optimal for mixing [31]; for a phase-separating mixture, the constant flow badly fails to homogenize the mixture.

VII. CONCLUSIONS

We have introduced the advective Cahn–Hilliard equation with a mean-zero driving term as a way of describing a stirred, phase-separating fluid, in the presence of sources and sinks. By specializing to symmetric mixtures, we have studied a more tractable problem, although one with many applications.

Our goal was to investigate stirring protocols numerically and analytically, and to determine the best way to break up the domains in the Cahn–Hilliard fluid and achieve homogenization. To this end, we introduced the m_p measure of composition fluctuations. Since in a well-mixed fluid, the composition exhibits spatial fluctuations about the mean, with better mixing leading to smaller fluctuations, we used m_p as a measure of mixedness or homogeneity. We proved the existence of m_p for long times, for $1 \leq p \leq 4$, and obtained *a priori* upper and lower bounds on m_4 , as an explicit function of the imposed flow $\mathbf{v}(\mathbf{x}, t)$, and the source $s(\mathbf{x})$.

We compared the level homogeneity achieved by the random-phase sine flow and the constant flow with the lower bound, and found that the sine flow is effective at homogenizing

the binary fluid, while the constant flow fails in this task. This is not surprising, since it is known that differential shears are needed to break up binary fluid domains, although it is radically different from the advection-diffusion case, where the constant flow was the optimal mixer. The question of whether or not a flow is a good mixer in this context was discussed using the mixing enhancement, defined in Sec. VI. Given such a definition, it is possible to compare stirring protocols and find the optimal protocol for mixing a binary fluid. Our upper bound on the enhancement provides a meaningful notion of this optimality. This result may be useful in applications where the homogenization of a binary fluid is desirable, since we have set a lower limit on precisely how much homogeneity can be achieved.

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- [1] A. Karim, J. F. Douglas, L. P. Sung, and B. D. Ermi. Self-assembly by phase separation in polymer thin films. *Encyclopedia of Materials: Science and Technology*, page 8319, 2002.
 - [2] D. G. A. L. Aarts, R. P. A. Dullens, and H.N.W. Lekkerherker. Interfacial dynamics in demixing systems with ultralow interfacial tension. *New J. Phys.*, 7:14, 2005.
 - [3] H. Wang and R. J. Composto. Thin film polymer blends undergoing phase separation and wetting: identification of early, intermediate, and late stages. *J. Chem. Phys.*, 113:10386, 2000.
 - [4] J. W. Cahn and J. E. Hilliard. Free energy of a nonuniform system. I. Interfacial energy. *J. Chem. Phys.*, 28:258–267, 1958.
 - [5] J. Lowengrub and L. Truskinowsky. Quasi-incompressible Cahn–Hilliard fluids and topological transitions. *Proc. R. Soc. Lond. A*, 454:2617–2654, 1998.
 - [6] D. S. Cohen and J. D. Murray. A generalized diffusion model for growth and dispersal in a population. *J. Math. Biology*, 12:237, 1981.
 - [7] C. M. Elliott and S. Zheng. On the Cahn–Hilliard equation. *Arch. Rat. Mech. Anal.*, 96:339–357, 1986.
 - [8] I. M. Lifshitz and V. V. Slyozov. The kinetics of precipitation from supersaturated solid solutions. *J. Chem. Phys. Solids*, 19:35–50, 1961.
 - [9] J. Zhu, L. Q. Shen, J. Shen, V. Tikare, and A. Onuki. Coarsening kinetics from a variable mobility Cahn–Hilliard equation: Application of a semi-implicit Fourier spectral method. *Phys. Rev. E*, 60:3564–3572, 1999.
 - [10] A. J. Bray. Theory of phase-ordering kinetics. *Adv. Phys.*, 43:357–459, 1994.
 - [11] S. Berti, G. Boffetta, M. Cencini, and A. Vulpiani. Turbulence and coarsening in active and passive binary mixtures. *Phys. Rev. Lett.*, 95:224501, 2005.
 - [12] L. Ó Náraigh and J.-L. Thiffeault. Dynamical effects and phase separation in thin films. *Phys. Rev. E*, *In press*, 2007.
 - [13] L. Berthier, J. L. Barrat, and J. Kurchan. Phase separation in a chaotic flow. *Phys. Rev. Lett.*, 86:2014–2017, 2001.
 - [14] L. Berthier. Phase separation in a homogeneous shear flow: Morphology, growth laws, and dynamic scaling. *Phys. Rev. E*, 63:051503, 2001.
 - [15] L. Ó Náraigh and J.L. Thiffeault. Bubbles and Filaments: Stirring a Cahn–Hilliard Fluid. *Phys. Rev. E*, 75:016216, 2007.

- [16] A. M. Lacasta, J. M. Sancho, and F. Sagues. Phase separation dynamics under stirring. *Phys. Rev. Lett.*, 75:1791, 1995.
- [17] A. J. Bray. Coarsening dynamics of phase-separating systems. *Phil. Trans. R. Soc. Lond.*, 361:781–792, 2003.
- [18] Z. Shou and A. Chakrabarti. Ordering of viscous liquid mixtures under a steady shear flow. *Phys. Rev. E*, 61:R2200, 2000.
- [19] T. Hashimoto, K. Matsuzaka, and E. Moses. String phase in phase-separating fluids under shear flow. *Phys. Rev. Lett.*, 74:126, 1995.
- [20] W. Lu and D. Salac. Patterning multilayers of molecules via self-organization. *Phys. Rev. Lett.*, 94:146103, 2005.
- [21] Z. Suo and W. Lu. Forces that drive nanoscale self-assembly on solid surfaces. *J. Nano. Res.*, 2:333, 2000.
- [22] L. Kielhorn and M. Muthukumar. Phase separation of polymer blend films near patterned surfaces. *J. Chem. Phys.*, 111:2259, 1999.
- [23] A. P. Krekhov and L. Kramer. Phase separation in the presence of spatially periodic forcing. *Phys. Rev. E*, 70:061801, 2004.
- [24] W. L. Craig, S. Danworaphong, and G. J. Diebold. Thermal diffusion in a sinusoidal temperature field. *Phys. Rev. Lett.*, 92:125901, 2004.
- [25] R. T. Pierrehumbert. Tracer microstructure in the large-eddy dominated regime. *Chaos, Solitons and Fractals*, 4:1091–1110, 1994.
- [26] P. Tong, W. I. Goldburg, J. Stavans, and A. Onuki. Temporal fluctuations in a turbulently stirred binary liquid mixture. *Phys. Rev. Lett.*, 62:2668, 1989.
- [27] C. R. Doering and J.-L. Thiffeault. Multiscale mixing efficiencies for steady sources. *Phys. Rev. E*, 74(2):025301(R), 2006.
- [28] T. A. Shaw, J.-L. Thiffeault, and C. R. Doering. Stirring up trouble: Multi-scale mixing measures for steady scalar sources. *Physica D*, 231(2):143–164, 2007.
- [29] J.-L. Thiffeault, C. R. Doering, and J. D. Gibbon. A bound on mixing efficiency for the advection–diffusion equation. *J. Fluid Mech.*, 521:105–114, 2004.
- [30] J.-L. Thiffeault and C. Doering. Multiscale mixing efficiency for steady sources. *Phys. Rev. E*, 74:025301(R), 2006.
- [31] J.-L. Thiffeault and G. Pavliotis. Optimizing the source distribution in fluid mixing. *arXiv:physics/0703135*, 2007.
- [32] P.V. Danckwerts. The definition and measurement of some characteristics of mixtures. *Appl. Sci. Res. A*, 3:279, 1952.
- [33] A. C. Edwards, W. D. Sherman, and R. E. Breidenthal. Turbulent mixing in tubes with transverse injection. *AIChE J.*, 31:516, 1985.
- [34] J. W. Cahn. Phase separation by spinodal decomposition in isotropic systems. *J. Chem. Phys.*, 42:93–99, 1965.
- [35] T. M. Antonsen, Jr., Z. Fan, E. Ott, and E. Garcia-Lopez. The role of chaotic orbits in the determination of power spectra. *Phys. Fluids*, 8(11):3094–3104, November 1996.
- [36] Z. Neufeld. Excitable media in a chaotic flow. *Phys. Rev. Lett.*, 87:108301, 2001.
- [37] R. T. Pierrehumbert. Lattice models of advection-diffusion. *Chaos*, 10:61–74, 2000.